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Experiments examining nonlinear scattering mechanisms of high intensity (2×10^{18} W/cm^2) laser light in underdense plasmas were performed. Red-shifted second harmonic emission at 45° from the directly backscattered direction was observed from field ionized plasmas having electron densities ranging up to 10^{19} cm^{-3}. This result was due to the doubling of stimulated Raman sidescattered light from the large plasma electron density gradients produced by ponderomotive cavitation.

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SECOND HARMONIC GENERATION OF STIMULATED RAMAN SCATTERED LIGHT IN UNDERDENSE PLASMAS

The development of short pulse high intensity laser systems [1] in the past several years has enabled researchers to explore new and sometimes unexpected physics which can occur in laser matter interactions as intensities approach 10^{19} W/cm^2. In this regime, conventional nonlinear optical theory is no longer valid [2] and, in fact, atoms in the target region will be ionized almost instantaneously by the leading edge of the laser field with subsequent interactions occuring in a plasma. Consequently, the role of the plasma on high intensity atomic physics phenomena [3] must be considered along with the effects of atomic and optical nonlinearities on the interaction between the laser pulse and the plasma. Proposed applications for high intensity lasers include electron acceleration, coherent x-ray generation, and laser "hole boring" - which has recently been postulated as a mechanism for enhancing the fusion rate in Inertial Confinement Fusion experiments [4]. In this scheme, a high intensity laser creates a channel through the outer "corona" plasma of a compressed deuterium-tritium pellet due to the large ponderomotive forces associated with the pulse. The interaction subsequently produces superthermal electrons which deposit energy into the center - thus igniting a sustained fusion reaction in the plasma.

At the Naval Research Laboratory high intensity laser scattering experiments were performed to elucidate some of the basic plasma physics phenomena which may be important for such applications. In these experiments a table top terawatt laser system was used which utilizes the technique of chirped pulse amplification [5] and which operates at a wavelength of 1.054 μm. The laser has a pulselength of 800 fsec, a maximum pulse energy of 1.2 J and can be focused to intensities greater than 2 x 10^{18} W/cm^2 using an f/3 off-axis parabolic mirror. At such intensities, underdense plasmas can be created due to field ionization of gas targets by the front of the laser pulse. Resulting plasma electrons in the laser field will have a significant mass increase from their relativistic "quiver" velocities (i.e. $\gamma = (1 + a_o^2)^{1/2} \approx 1.5$ where $a_o = v_{osc}/c$).

The scattered radiation emitted from plasmas produced by the ionization of a pulsed jet of hydrogen or helium gas was examined by imaging the light onto the entrance slit of a spectrometer operating in the visible and near infrared region. Output from the spectrometer was

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directed into a streak camera having 10 psec resolution with a sweep duration of 0.5 nsec. The radiation from plasma ion recombination in these experiments occurs on a much longer timescale than the streak camera sweep and was not of interest, so only light emitted at times close to the interaction of the laser with the target gas jet (i.e. within 50 psec) was measured. Scattered light was collected by a lens having an acceptance angle of 22° and the spectrum was examined at various angles with respect to the incident laser direction as well as for various electron densities and laser intensities. The plasma electron density in the ionized gas jet (10^{18} - 10^{19} cm^{-3}) was inferred to within approximately 25% from observations of the backscattered Raman spectrum from experiments at relatively low intensities (10^{16} W/cm^2).

At a 45° angle from the directly backscattered direction, spectra were observed which exhibit a narrow peak close to the incident laser frequency and a surrounding broad oscillatory structure extending into the long and short wavelength regions around the fundamental. This broad structure (~ 1200 Å wide) is qualitatively similar to the spectrum of stimulated Raman backscatter (SRS) [6] and stimulated Brillouin backscatter (SBS) [6] which have been previously observed in the directly backscattered direction [7] and is probably produced by similar mechanisms. The sidescattered spectral structure is not as broad as that observed in the directly backscattered direction and the signal is several orders of magnitude weaker.

When the sidescattered spectrum at 45° is examined near the second harmonic frequency a broad red-shifted feature is observed. Two scenarios are plausible for the generation of a red-shifted second harmonic signal : i) laser light may be frequency doubled as it propagates through the interaction region and subsequently redshifted due to stimulated Raman sidescattering in the plasma - leading to a shift of ω_{pe} (the plasma frequency) - or, ii) laser radiation may be first Raman sidescattered (red shifted by ω_{pe}) and only frequency doubled as it passes out of the plasma - resulting in an apparent frequency shift of $2\omega_{pe}$. From Fig. 1a, it is clear that the observed red shift in the signal measured near the second harmonic is approximately $2\omega_{pe}$ and that the second process is occuring, i.e., $(\omega + \omega_{pe}) + (\omega + \omega_{pe}) \rightarrow 2\omega + 2\omega_{pe} \rightarrow \omega_{measured}$.

There have been predictions of the production of sidescattered second harmonic radiation - in particular nonlinear Thomson scattering [8] due to relativistic effects. Theory and computer simulations of this mechanism for laser intensities of 2 x 10^{18} W/cm^2 also predict comparable levels of harmonic emission at other angles and at higher orders - however none were observed. In the experiments described here, emission in the second harmonic region was also measured at relatively low laser intensities of 2 x 10^{17} W/cm^2 where relativistic effects should be negligible. As shown in Fig. 1b, in these experiments, the scaling of signal strength vs. density is clearly nonlinear in contrast to relativistic Thomson sidescattering which should show a linear scaling with electron density.

In Fig. 2, 45° second harmonic spectra are shown for various peak laser intensities at an electron density of about 10^{19} cm^{-3}, indicating that the broadening varied with laser intensity. The signal was approximately 400 nm wide at maximum laser intensity and exhibited an oscillatory structure similar to that shown in the sidescattered fundamental. The spectra also showed significant blueshifting as the intensity of the laser light increased. This is similar to the behavior of the directly backscattered SRS light which also blueshifts as the laser intensity increases and which has been observed previously [7]. In the experiments described here, however, the amount of blueshifting seems to saturate at intensities above 10^{18} W/cm^2.

Changing the incident laser polarization from linear to circular had little effect on the spectra. There was also no emission in the third harmonic region. No significant emission in the visible and near infrared spectrum was observed at a 90 degree angle using the same detection system. These observations indicate the absence of harmonic generation by atomic processes, relativistic effects and filamentation.

Even though there are significant amounts of stimulated Raman scattered light in the directly backscattered direction a second harmonic feature of this type was not observable there. In fact, at intensities of 2 x 10^{18} W/cm^2 the SRS spectrum in the directly backscattered direction is broadened to extend beyond the second harmonic (to about 450 nm) [9]. However, in the

region to the long wavelength side of 527 nm a "hole" occurs in the spectrum - appearing as if an absorption process is occuring (see Fig. 3).

It is likely that emission of the second harmonic of the sidescattered radiation is due to propagation of the the stimulated Raman sidescattered light across the steep density gradients surrounding a region of cavitation in the gas jet. For very high intensity light a region of "cavitation" [11] can be caused by the expulsion of electrons due to the tremendous ponderomotive force of the focused laser light while ions are approximately stationary since they are much more massive. The ponderomotive potential felt by the plasma electrons is given by $\Phi = e^2E^2/4m_e\omega_o^2$, where **E** is the electric field of the laser, ω_o is the laser frequency and m_e is the electron mass and was 170 keV in this experiment for laser pulses at maximum intensity. Thus, significant charge separation can occur and a cylindrical shell of excess electrons which have been expelled from the interaction region is formed. The amount of cavitation can be simply estimated by a force balance between the ponderomotive force of the laser pulse and the space charge field. In this case, if r_o is the laser spot size the density perturbation is given approximately by $n_1 = \Phi \ / \ (4\pi e r_o^2 \ )$ which is about 2 x 10^{17} cm^{-3} for the laser at maximum intensity. This is about 2 - 10 % of the electron densities used in this experiment. It should be noted that if self-focusing occurs and increases the ponderomotive field a larger density perturbation results.

The second harmonic generation process can be described by the solution of the wave equation with a source term due to the nonlinear current produced as the scattered light passes through the density gradient [10], i.e.,

$$(\nabla^2 - k^2) \, a_2 = k_p^2 \frac{n_1}{n_o} a_1$$

where n_o is the plasma electron density, k_p (= c ω_{pe}) is the plasma wavenumber, a_1 is the normalized laser intensity of the stimulated Raman sidescattered light and a_2 is the doubled sidescattered light. From this equation it can be shown that the efficiency of the second order harmonic generation process is given by;

$$\frac{P_2}{P_1} = a_1^2 \left(\frac{L}{4L_p} \frac{k_p^2}{k_1^2} \right)^2$$

where L_p is the density gradient scalelength, L is the interaction distance, and k_1 is the wavenumber of the scattered laser light. Using parameters from the present experiment the predicted efficiency of this process is approximately $10^{-8\pm1}$. The experimental conversion efficiency was not known precisely but if reasonable assumptions are used to estimate the signal strength it appears that the measured value is a couple orders of magnitude greater than this. This may be due to errors in the estimations used and the fact that self-focusing in the plasma may produce density gradients scalelengths which are less than the focal spot size.

There are other sharp plasma density gradients in the interaction region especially that at the boundary where the gas jet is ionized - although the intensity of the sidescattered light may be too low at this boundary to produce significant doubling. As well, the three-body recombination time for cold, dense plasmas can by quite short (i.e. a few picoseconds) implying that this gradient may be reduced as the plasma quickly recombines. Harmonic generation is also critically dependent upon phase matching conditions in the medium [12]. In a plasma free electrons cause a positive dispersion between the fundamental and the harmonic and one can approximate the detuning length for the harmonic generation process. The plasma refractive index is given by $\eta=[1-(\omega_p/\omega_0)^2]^{1/2}$, and the corresponding detuning length is

$$L_d = \frac{\pi}{\Delta k} = \frac{\pi c}{q\omega_o \Delta\eta} = \frac{2\pi q c \omega}{\omega_{pe}^2 (q^2 - 1)}$$

which becomes significantly shorter with increasing harmonic order. For the second harmonic this detuning length is approximately 70 μm which is much longer than the density gradient scalelength (~ the laser spot size ~ 10 μm) produced by the cavitation process .

The lack of a second harmonic signal in the directly backscattered direction may be also due to this effect. The plasma gradient should be smaller in the backward direction - reducing the conversion efficiency - and, the interaction distance should be either the laser pulse width or the Rayleigh length - whichever is less. Here the Rayleigh length is 250 μm and is greater than the

detuning distance in this case. Large amplitude plasma wakefields may also be produced which may increase the fluctuation levels in the plasma and prevent good phase matching. It is, however, difficult to see how phase mismatches might result in the "dip" in the backscattered spectrum near the second harmonic wavelength or even how unionized hydrogen atoms might exist in the path of the backscattered Raman light to provide some sort of atomic absorption mechanism. Second harmonic generation has been previously observed at a 90° angle to the direction of the laser propagation during interactions of much lower intensity light (10^{14} - 10^{15} W/cm^2) with preformed near-critical density plasmas [13]. This emission was associated with the filamentation instability and an interaction between the forward traveling fundamental and a backscattered wave due to SBS - in contrast to the experiments described here where sidescattered light is doubled.

The production of second harmonic radiation can also occur from atomic processes - however this requires the existence of a spatial asymmetry since, for isotropic media such as gases or liquids, generation of even harmonics can not occur due to atomic parity considerations [12]. In this experiment, such a spatial asymmetry can be caused by an instantaneous radial static electric field produced in the cavitation process. However, for hydrogen, in the region where significant static electric fields exist the focused intensity is very high and there are no unionized atoms to be polarized. In the region where neutral atoms exist (i.e where the laser intensity never exceeds about 10^{14} W/cm^2) the static electric field is very small.

Previous work has observed second harmonic generation in the forward direction using metal vapors where the mechanism of charge separation was postulated [14] - and work with gas jet targets has also observed second harmonic generation in the forward direction (at laser intensities up to 10^{16} W/cm^2) due to this effect [15]. The efficiency observed ($\sim 10^{-14}$) in those experiments was found to be two orders of magnitude less than that for generation of the third harmonic. In experiments described here, spectra were also recorded for forward scattered light at laser intensities of 2×10^{18} W/cm^2, and the emission of the second harmonic (with a relatively broad bandwidth of 7 nm) was found to be strong for helium gas targets (with an

efficiency of ~ $10^{-6\pm1}$) and was probably due to such atomic effects. No significant signal from the third harmonic was measured. The magnitude of the static radial electric field produced by the cavitation process can be simply estimated [15] by $\mathbf{E}_s = \mathbf{\Phi} / er_0$ - giving a value of 2 x 10^8 V/cm (note that the value of the maximum laser electric field is ~ 3 x 10^{10} V/cm). The second harmonic signal was also observed, in our experiments, to be blue-shifted by 120 Å (much larger than the average blueshift of the forward scattered fundamental) indicating that such harmonics are produced only at the leading edge of the laser pulse where the ionization rate of the plasma is the highest. Second harmonic emission is thus produced by the singly ionized helium atoms which remain in the laser electric field but which are also subject to an instantaneous static electric field produced by cavitation. In contrast, significant second harmonic generation was not observed for hydrogen gas. This was expected since in hydrogen - after initial ionization and charge separation - there are no neutral atoms remaining to be polarized which could emit harmonics in the forward direction.

In conclusion, these experiments have demonstrated that cavitation effects are taking place which result in large density gradients in the laser plasma interaction region capable of producing significant second harmonic radiation. These observations provide the first measurements of frequency doubling of stimulated Raman scattered laser light as well as the first measurements of frequency doubling of intense laser light caused by very underdense, fully ionized plasma phenomena. At these intensities, there is clearly a complex interrelation between the mechanisms of second harmonic generation, cavitation and stimulated Raman scattering. In the experiments discussed here the ponderomotive force should not be strong enough to push aside ions and create a "hole" in the plasma - however such effects may occur for higher intensity lasers and the phenomena observed in these experiments may become even more pronounced. In fact, the measurement of the angular distribution of second harmonic backscatter may provide a useful diagnostic of "hole-boring" in ICF experiments which utilize the "Fast Igniter" concept.

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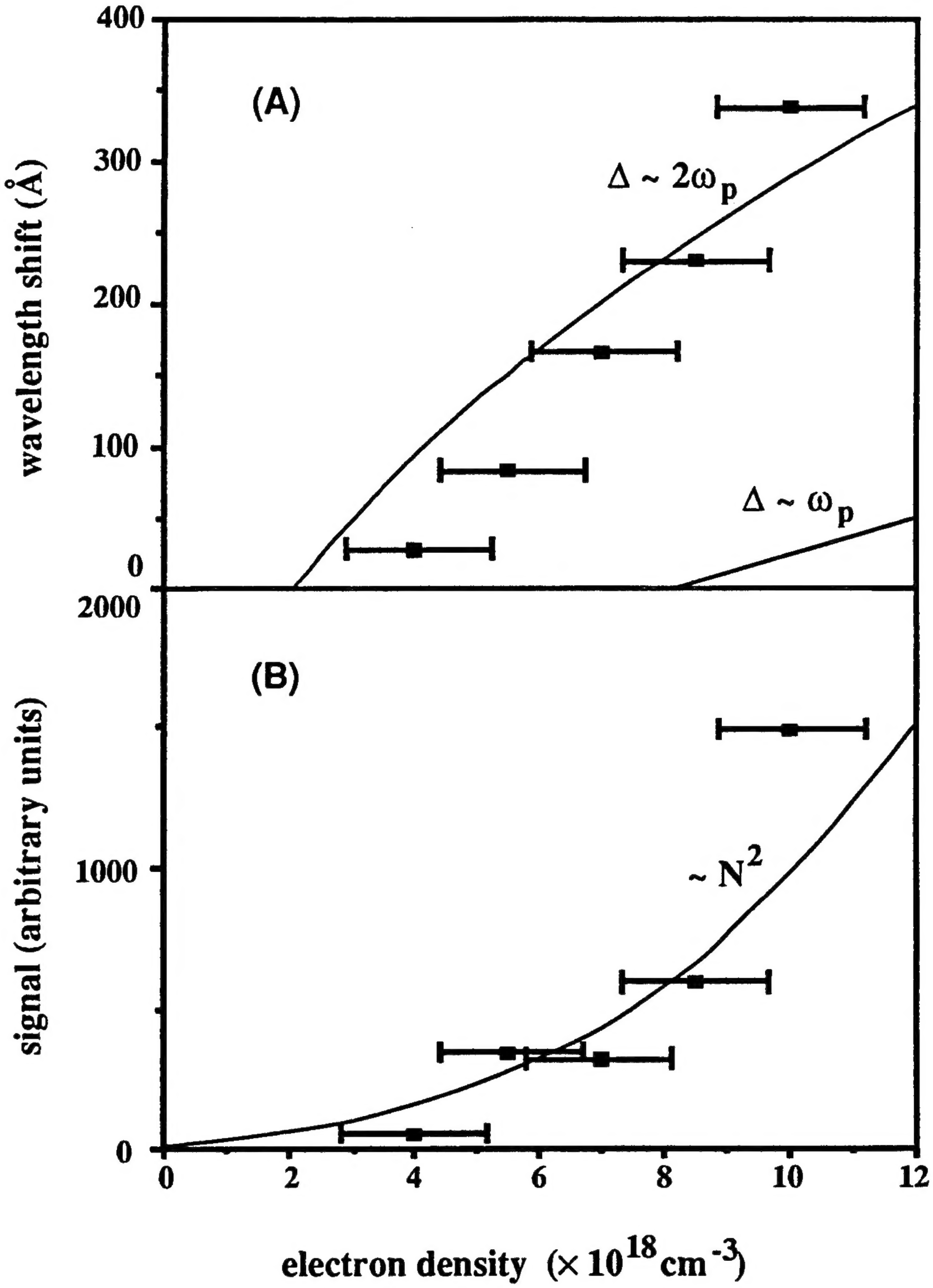

FIG. 1 A) Wavelength shift of sidescattered second harmonic emission vs. electron density (solid lines correspond to shifts of ω_p and $2\omega_p$ using 250 Å intensity dependent blueshift), B) Plot of second harmonic sidescatter emission intensity vs. electron density (incident laser intensity was ~ 1.0×10^{18} W/cm^2).

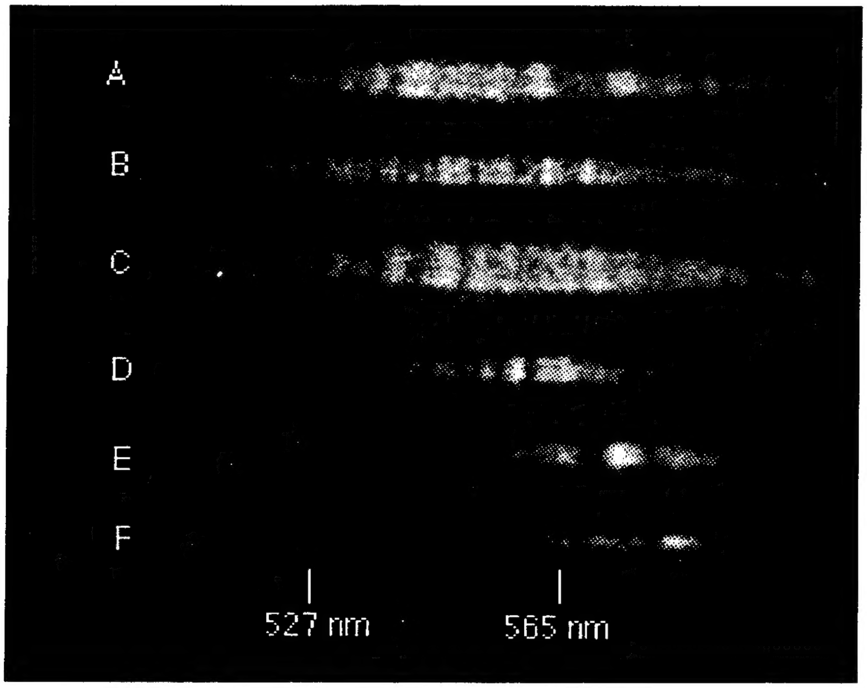

FIG. 2 Variation of second harmonic at 45° backscattered direction with respect to incident laser intensity: A) 2.0×10^{18} W/cm^2, B) 1.5×10^{18} W/cm^2, C) 1.0×10^{18} W/cm^2, D) 6×10^{17} W/cm^2, E) 5×10^{17} W/cm^2, and F) 3×10^{17} W/cm^2.

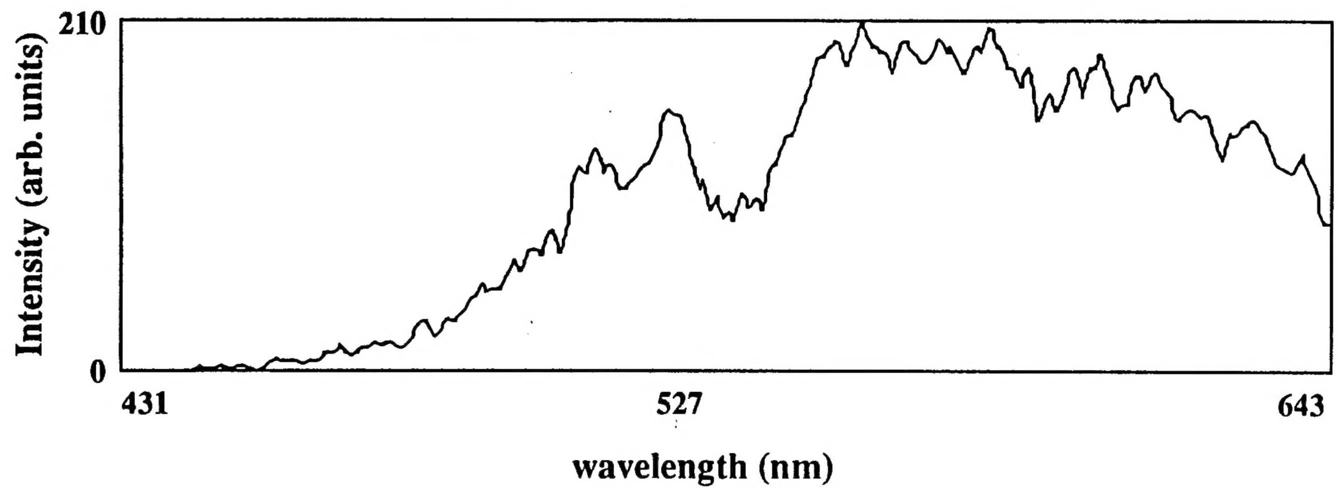

FIG. 3 "Dip" in directly backscattered spectrum in region of second harmonic.